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ABSOLUTE CALIBRATION OF A URANIUM - 238 FAST-NEUTRON DETECTOR. (U)

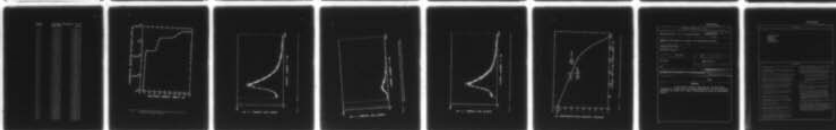
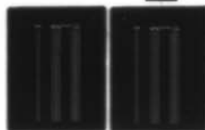
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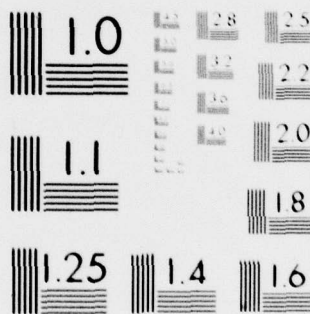
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6 ABSOLUTE CALIBRATION OF A URANIUM - 238  
FAST-NEUTRON DETECTOR

by

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### ABSTRACT

A fast-neutron detector, employing the neutron-induced fissioning of uranium - 238 as the detecting mechanism, has been calibrated absolutely for future use at DREO in monitoring the output of fast-neutron sources.



### RÉSUMÉ

Un détecteur à neutrons rapides qui utilise comme mécanisme de détection la fission de  $^{238}\text{U}$  induite par les neutrons, a été calibré de façon absolue pour l'utilisation future au CRDO pour le contrôle de sources à neutrons rapides.

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## INTRODUCTION

Proportional radiation detectors employing fissionable materials as the particle-detecting isotope are called "fission chambers". Such detectors typically contain small amounts of fissionable material plated on the inner surfaces of an otherwise normal gas chamber, operated in the proportional-counter region of applied high voltage. Neutron-induced nuclear fission results in the breakup of the fissioning nucleus into two energetic fission fragments, travelling in mutually opposite directions. If the plating thickness is significantly less than the range of the fission fragments in the plating material, then one fragment will escape into the fill gas of the counter. As the fragment typically has about 80 MeV of kinetic energy and a net charge of the order of 21, considerable ionization is created in the gas during the slowing down of the particle. In the manner normal for proportional gas counters, this ionization is collected by an applied high voltage gradient within the chamber, and following suitable amplification, results in a recognizable electronic pulse.

For the particular case of a chamber containing uranium - 238 with a plating thickness of about  $0.5 \text{ mg/cm}^2$ , the device offers certain advantages over other detecting systems when used to detect neutrons whose energies exceed 1.5 MeV. Since the number of ions created by the recoiling fission fragments is more than would be created by gamma rays traversing the detector, discrimination above a certain output pulse voltage removes all signals due to gamma rays. For sufficiently thin plating, most of the fission fragment's kinetic energy will be deposited in the fill gas; consequently the pulse-height spectrum will show a well resolved peak. This peak will occur at a voltage considerably above that expected from sources of electronic noise. As uranium -238 itself is alpha-radioactive, the detector contains its own radiation check source, which may be observed if the electronic discrimination level is reduced sufficiently to allow detection of the ionization resulting from the emitted alpha particles. This characteristic is useful in verifying the correct operation of the associated electronic equipment. The effective threshold of the (n,f) reaction of uranium - 238 occurs at approximately 1.5 MeV of neutron energy, consequently the detector is essentially insensitive to neutrons of lower energy. In experimental situations where the source or detector are in close proximity to extraneous scattering materials, this threshold will tend to reduce the fraction of observed neutrons which have been scattered to lower energies, thus indicating with greater precision the actual output of the neutron source.

The major disadvantage of the detector results from its small efficiency, due to the low (n,f) cross-section and the necessity of using very thin material plating. However, if the detector is employed to monitor the output of intense fast-neutron sources, this relative inefficiency may be beneficial, in that even in high radiation fields the gradual burn-up of detecting material will be insignificant and no dead-time losses are incurred. In addition, the detector may be located quite close to the neutron source, thus by geometric considerations, further reducing the effective scattered-neutron sensitivity, without resulting in an excessively high observed neutron-counting rate.

Finally, as the uranium - 238 (n,f) reaction has been studied intensively, its energy-dependent cross section is known to a high degree of accuracy. If the amount of uranium - 238 contained in the detector is also known, then the absolute, energy-dependent detection efficiency may be calculated accurately from first principles.

#### THEORETICAL DETECTION EFFICIENCY

Since the detector is essentially "white" to incident fast neutrons, self-shielding of the detecting material will be negligible. Consequently, the detection efficiency is given simply by:

$$\eta(E) = N_{238} \sigma_f(E) \quad (1)$$

where:  $\eta(E)$  is the energy-dependent efficiency or sensitive area ( $\text{cm}^2$ )

$N_{238}$  is the total number of uranium - 238 nuclei present

$\sigma_f$  is the energy-dependent fission cross section ( $\text{cm}^2$ )

$E$  is the incident neutron energy (MeV)

A recent evaluation by Ing and Cross<sup>1</sup>, has shown that the (n,f) cross section of uranium - 238 may be represented to within  $\pm 6\%$  by:

$$\begin{aligned} \sigma_f(E) = 1.33 - \frac{0.51}{1 + (E/1.5)^{11}} - \frac{0.45}{1 + (E/6.4)^{20}} \\ - \frac{0.37}{1 + (E/14.1)^{20}} + 0.03 \exp[-(3E - 6)^2] \end{aligned} \quad (2)$$

The presence of other fissionable isotopes of uranium, such as  $^{235}\text{U}$  may be ignored, provided the amount present relative to  $^{238}\text{U}$  is sufficiently small.

The fission counter currently used at DREO (Reuter-Stokes Model RS-P6-1608-112, Serial number 4-3268) has a uranium - 238 coating in the chemical form  $\text{U}_3\text{O}_8$ . The uranium was prepared and assayed<sup>2</sup> by the Oak Ridge National Laboratory (U.S.A.), with the following results:

$^{233}\text{U}$	<1 ppm
$^{236}\text{U}$	<1 ppm
$^{235}\text{U}$	22 ppm
$^{236}\text{U}$	<1 ppm
$^{238}\text{U}$	99.9975%



The presence of  $^{235}\text{U}$  will not change appreciably the fast-fission cross section of the mixture (approximately 0.005%), while the thermal-neutron cross section will still be less than 1% compared to that at 3 MeV.

The manufacturer claims<sup>3</sup> that the  $\text{U}_3\text{O}_8$  was coated to a thickness of  $0.5969 \text{ mg/cm}^2 \pm 3.7\%$ , over a total plated area of  $1000 \text{ cm}^2$ .

Thus, the total number of uranium - 238 nuclei present in the counter is calculated as:

$$N_{238} = 0.5969 \frac{\text{mg}}{\text{cm}^2} \times 1000 \text{ cm}^2 \times \frac{(3 \times 238)}{(3 \times 238) + (8 \times 16)} \times \frac{6.02 \times 10^{23}}{238 \text{ g}} \times \frac{1 \text{ g}}{1000 \text{ mg}}$$

$$N_{238} = 1.28 \times 10^{21} \text{ nuclei} \pm 3.7\%$$

The zero bias efficiency  $\eta_0(E)$ , (i.e., assuming all fission events are recorded by the counting electronics) is thus given by the product of this value with equation (2), as in equation (1). For the sake of convenience, calculated zero-bias efficiencies are listed in Appendix A for neutron energies from 0 to 20 MeV, in 0.1-MeV increments, and also shown plotted in Figure 1.

#### EXPERIMENTAL DETECTION EFFICIENCY

In order to determine experimentally the efficiency of an arbitrary detector, a radioactive source of known spectral shape and calibrated total intensity is required. For the purpose of calibrating fast-neutron monitors, neutrons emitted during the spontaneous fission of californium - 252 are ideal in that the neutron spectrum is known accurately, and the intensity of neutrons produced per gram of the material is sufficiently high to render insignificant the perturbation of the neutron spectrum due to self-shielding of the source by itself. Such a source was recently made available to DND by the United States Department of Energy and was used, among other projects, to calibrate experimentally the  $^{238}\text{U}$  counter.

The detection efficiency of the counter in measuring  $^{252}\text{Cf}$  source neutrons may be written:

$$\langle \eta \rangle = \frac{\int_0^\infty N^{238} \sigma_f(E) \phi(E) dE}{\int_0^\infty \phi(E) dE} \quad (3)$$

which is merely a calculation of the mean detection efficiency, averaged over the spectrum of source neutrons.

The spectrum of californium - 252 neutrons has been determined by several researchers<sup>4</sup> to be given by a Maxwellian flux spectrum, at an

effective neutron temperature  $kT = 1.45$  MeV. Thus:

$$\phi(E) = \frac{\sqrt{E} e^{-E/kT}}{(kT)^{3/2}} \quad (4)$$

Substituting the spectrum (4) into equation (3), and performing the integrals by numerical methods, yields:

$$\langle \eta \rangle = N_{238} \langle \sigma_f \rangle \quad (5)$$

with a value of 0.302 barns ( $\pm 6\%$ ) calculated for  $\langle \sigma_f \rangle$ .

The experimental efficiency,  $\eta$ , is determined by placing the source apart from the detector a known distance, recording the observed counting rate, and comparing that to the neutron flux at the detector calculated using the known source intensity.

From the original assay<sup>5</sup> of the neutron emission rate, and correcting for source decay since the date of assay, yields a present (21 June 78) source strength of:

$$S_0 = 3.855 \times 10^9 \text{ n/sec} \pm 3\% \quad (6)$$

Correcting for the attenuation of neutrons in passing through the source holder (1/8" aluminium) and 2.43 metres of air, yields an effective source strength of:

$$S = S_0 \times 0.9985 \times 0.980 = 3.72 \times 10^9 \text{ n/sec} \pm 3\% \quad (7)$$

At the detector position (2.43 metres distant from the source), the total uncollided neutron flux is thus:

$$\phi_T = \frac{S}{4\pi r^2} = 5016 \text{ neutrons/cm}^2\text{-sec} \pm 3.1\%$$

the quoted error reflecting both the uncertainty in the original source assay (3%) and the source-to-detector distance (0.41%).

Pulse-height signals, after amplification by a CI<sup>6</sup> 806 pre-amplifier (at a high-voltage bias of 600 volts) and a CI 814 main amplifier (net gain = +40) were analysed into 1024 channels of pulse height by a CI 8180 multi-channel analyser, for a total live-time period of 40,000 seconds. The observed spectrum is shown in figure 2, with the fission-fragment peak occurring at channel 280, and the alpha-particle contribution falling below channel 115. Note that to reduce degradation of pulse-height resolution, the analyser's lower-level discriminator was adjusted such that the ADC dead time was below 2%, corresponding to a lower-level threshold at channel 40. From figure 2, it is seen that the minimum differential counting rate occurs at channel 141, corresponding to the apparent minimum in the "valley" between alpha-particle and fission-fragment initiated signals. For single-channel counting

purposes, such as in monitoring total output of variable neutron sources, lower-level discrimination at this point will result in the greatest integral stability and immunity against possible high-voltage, amplifier, and discriminator drifting. Conveniently this valley occurs at exactly half of the fission-fragment peak pulse height, and thus may be found easily using only single-channel counting systems. Note that discrimination at this point will also eliminate any alpha-particle counting of significance.

In order to measure the contribution of scattered neutrons to the observed spectrum, a second measurement was made with a 16" long paraffin cylinder interposed between the source and detector. This cylinder effectively scatters all uncollided neutrons which would otherwise intercept the detector, while causing minimal perturbation of the scattered contribution. The spectrum of pulse heights thus measured is shown in figure 3. Subtracting channel by channel from the first measurement yields the spectrum of events due only to the uncollided component and is shown in figure 4. The integral of this spectrum represents the true counting rate due to uncollided neutrons, less that fraction of pulse heights falling below channel 110, which are effectively masked by the inherent alpha-particle contribution. To estimate the "zero-bias" integral counting rate, it is necessary to extrapolate to channel zero, the observed integral pulse-height distribution, which is formed by summing sequentially the differential spectrum above each channel. Figure 5 represents the low-pulse-height portion of the integral distribution so calculated. Below channel 200, the experimental curve is linear, with a linearity correlation coefficient of 0.997 and an extrapolated zero-bias value of  $2.031 \pm 0.008$  counts per second.

$$\therefore \eta_e = \frac{C_0}{\phi_T} = \frac{2.031 \pm 0.008}{5016 \pm 156} = 4.05 \times 10^{-4} \text{ cm}^2 \quad (\pm 3.2\%)$$

The corresponding theoretical zero-bias efficiency is given by equation (5) and the value of  $N_{238}$  determined previously, i.e.,

$$\begin{aligned} \eta_t &= N_{238} \langle \sigma_f \rangle = (1.28 \times 10^{21} \pm 3.71\%) (0.302 \times 10^{-24} \pm 6\%) \\ &= 3.87 \times 10^{-4} \text{ cm}^2 \quad (\pm 7.1\%) \end{aligned}$$

The difference being equal to  $0.18 \times 10^{-4} \text{ cm}^2$  with an expected error in this value of  $0.39 \times 10^{-4} \text{ cm}^2$ . Since this difference corresponds to less than one standard error, it must be concluded that the calculated and experimental efficiencies do not differ significantly.



### RECOMMENDATIONS

The recommended zero-bias efficiency curve for future use of the detector is consequently that predicted by theoretical calculation. If the counting apparatus is biased according to the previously described procedure, i.e., lower-level discrimination at a pulse height equal to one-half of the peak fission-fragment pulse height, then the biased efficiency will be reduced by a factor of 0.894 (see figure 5). The total, biased efficiency will thus be given by 0.894 times the predicted zero-bias efficiency; and is listed numerically in Appendix A. Under these conditions, the integral discrimination stability will be 0.13, or a 1.3% change in counting rate for a 10% change in effective discrimination level (from the linear slope of figure 5). Furthermore, total immunity against alpha-particle counting will be assured provided the discrimination level does not drift downwards by more than 21% of the set value. The efficiencies listed in Appendix A may be considered accurate to within a standard error of  $\pm 6\%$ .

REFERENCES

1. "Analytical Representation of Some Fast-Neutron Cross Sections useful in Dosimetry"; W.G. Cross and H. Ing; Nucl. Sci. Engrg., 58, 377-386 (1975).
2. Private communication;  $^{238}\text{U}$  Assay, Oak Ridge National Laboratory, Isotope Order No. 29-2526 (1976).
3. Private communication; Reuter Stokes Ltd., (28 Oct. 1976).
4. "Fission Neutron Spectrum of  $^{252}\text{Cf}$ "; J. Csikai and Z. Dezso, Annals of Nuclear Energy; 3, 527-530 (1976).
5. Private Communication:  $^{252}\text{Cf}$  Assay, Savannah River Laboratory, Source Identification No. SR-Cf-318C, (20 Oct. 1977)
6. Canberra Industries Ltd., Meriden, Connecticut.



## APPENDIX A

<u>ENERGY</u> (MEV)	<u>DETECTOR EFFICIENCY (CM**2)</u>	
	<u>ZERO BIAS</u>	<u>BIASED</u>
.1	.000E 00	.000E 00
.2	.000E 00	.000E 00
.3	.000E 00	.000E 00
.4	.000E 00	.000E 00
.5	3.662E-09	3.274E-09
.6	2.686E-08	2.401E-08
.7	1.489E-07	1.331E-07
.8	6.482E-07	5.795E-07
.9	2.351E-06	2.111E-06
1.0	7.456E-06	6.674E-06
1.1	2.087E-05	1.866E-05
1.2	5.176E-05	4.627E-05
1.3	1.125E-04	1.006E-04
1.4	2.097E-04	1.874E-04
1.5	3.304E-04	2.954E-04
1.6	4.467E-04	3.994E-04
1.7	5.333E-04	4.813E-04
1.8	6.022E-04	5.383E-04
1.9	6.428E-04	5.746E-04
2.0	6.647E-04	5.943E-04
2.1	6.722E-04	6.009E-04
2.2	6.701E-04	5.990E-04
2.3	6.640E-04	5.936E-04
2.4	6.582E-04	5.884E-04
2.5	6.545E-04	5.851E-04
2.6	6.528E-04	5.836E-04
2.7	6.523E-04	5.831E-04
2.8	6.522E-04	5.831E-04
2.9	6.524E-04	5.832E-04
3.0	6.525E-04	5.833E-04
3.1	6.526E-04	5.834E-04
3.2	6.526E-04	5.835E-04
3.3	6.527E-04	5.835E-04
3.4	6.527E-04	5.835E-04
3.5	6.527E-04	5.836E-04
3.6	6.528E-04	5.836E-04
3.7	6.528E-04	5.836E-04
3.8	6.528E-04	5.836E-04
3.9	6.528E-04	5.836E-04
4.0	6.528E-04	5.836E-04
4.1	6.529E-04	5.837E-04
4.2	6.529E-04	5.837E-04
4.3	6.530E-04	5.838E-04
4.4	6.531E-04	5.839E-04
4.5	6.533E-04	5.840E-04
4.6	6.536E-04	5.843E-04
4.7	6.540E-04	5.847E-04
4.8	6.546E-04	5.852E-04
4.9	6.555E-04	5.861E-04
5.0	6.569E-04	5.873E-04

<u>ENERGY</u> (MEV)	<u>DETECTOR EFFICIENCY (CM**2)</u>	
	<u>ZERO BIAS</u>	<u>BIASED</u>
5.1	6.539E-04	5.390E-04
5.2	6.617E-04	5.916E-04
5.3	6.658E-04	5.952E-04
5.4	6.714E-04	6.003E-04
5.5	6.793E-04	6.073E-04
5.6	6.901E-04	6.169E-04
5.7	7.045E-04	6.298E-04
5.8	7.234E-04	6.467E-04
5.9	7.474E-04	6.682E-04
6.0	7.771E-04	6.947E-04
6.1	8.123E-04	7.262E-04
6.2	8.523E-04	7.620E-04
6.3	8.958E-04	8.009E-04
6.4	9.408E-04	8.411E-04
6.5	9.851E-04	8.807E-04
6.6	1.027E-03	9.179E-04
6.7	1.064E-03	9.514E-04
6.8	1.097E-03	9.805E-04
6.9	1.124E-03	1.005E-03
7.0	1.147E-03	1.025E-03
7.1	1.165E-03	1.041E-03
7.2	1.179E-03	1.054E-03
7.3	1.190E-03	1.064E-03
7.4	1.199E-03	1.072E-03
7.5	1.206E-03	1.073E-03
7.6	1.211E-03	1.083E-03
7.7	1.215E-03	1.086E-03
7.8	1.218E-03	1.089E-03
7.9	1.220E-03	1.091E-03
8.0	1.222E-03	1.093E-03
8.1	1.224E-03	1.094E-03
8.2	1.225E-03	1.095E-03
8.3	1.226E-03	1.096E-03
8.4	1.226E-03	1.096E-03
8.5	1.227E-03	1.097E-03
8.6	1.227E-03	1.097E-03
8.7	1.228E-03	1.097E-03
8.8	1.228E-03	1.098E-03
8.9	1.228E-03	1.098E-03
9.0	1.228E-03	1.098E-03
9.1	1.228E-03	1.098E-03
9.2	1.228E-03	1.098E-03
9.3	1.229E-03	1.098E-03
9.4	1.229E-03	1.098E-03
9.5	1.229E-03	1.099E-03
9.6	1.229E-03	1.099E-03
9.7	1.229E-03	1.099E-03
9.8	1.229E-03	1.099E-03
9.9	1.229E-03	1.099E-03
10.0	1.229E-03	1.099E-03

<u>ENERGY</u> (MEV)	<u>DETECTOR EFFICIENCY (CM**2)</u>	
	<u>ZERO BIAS</u>	<u>BIASED</u>
10.1	1.229E-03	1.099E-03
10.2	1.229E-03	1.099E-03
10.3	1.230E-03	1.099E-03
10.4	1.230E-03	1.099E-03
10.5	1.230E-03	1.100E-03
10.6	1.230E-03	1.100E-03
10.7	1.231E-03	1.100E-03
10.8	1.231E-03	1.101E-03
10.9	1.232E-03	1.101E-03
11.0	1.232E-03	1.101E-03
11.1	1.233E-03	1.102E-03
11.2	1.233E-03	1.103E-03
11.3	1.234E-03	1.104E-03
11.4	1.235E-03	1.104E-03
11.5	1.237E-03	1.106E-03
11.6	1.238E-03	1.107E-03
11.7	1.240E-03	1.108E-03
11.8	1.242E-03	1.110E-03
11.9	1.244E-03	1.112E-03
12.0	1.247E-03	1.115E-03
12.1	1.250E-03	1.118E-03
12.2	1.254E-03	1.121E-03
12.3	1.258E-03	1.124E-03
12.4	1.252E-03	1.129E-03
12.5	1.263E-03	1.133E-03
12.6	1.274E-03	1.139E-03
12.7	1.281E-03	1.145E-03
12.8	1.289E-03	1.152E-03
12.9	1.297E-03	1.160E-03
13.0	1.307E-03	1.168E-03
13.1	1.317E-03	1.178E-03
13.2	1.329E-03	1.188E-03
13.3	1.341E-03	1.199E-03
13.4	1.354E-03	1.211E-03
13.5	1.369E-03	1.224E-03
13.6	1.384E-03	1.237E-03
13.7	1.399E-03	1.251E-03
13.8	1.415E-03	1.265E-03
13.9	1.432E-03	1.280E-03
14.0	1.449E-03	1.295E-03
14.1	1.466E-03	1.310E-03
14.2	1.482E-03	1.325E-03
14.3	1.499E-03	1.340E-03
14.4	1.515E-03	1.354E-03
14.5	1.530E-03	1.368E-03
14.6	1.545E-03	1.381E-03
14.7	1.559E-03	1.394E-03
14.8	1.572E-03	1.405E-03
14.9	1.584E-03	1.416E-03
15.0	1.596E-03	1.427E-03

<u>ENERGY</u> (MEV)	<u>DETECTOR EFFICIENCY (CM*2)</u>	
	<u>ZERO BIAS</u>	<u>BIASED</u>
15.1	1.606E-03	1.436E-03
15.2	1.616E-03	1.445E-03
15.3	1.625E-03	1.453E-03
15.4	1.633E-03	1.460E-03
15.5	1.640E-03	1.467E-03
15.6	1.647E-03	1.472E-03
15.7	1.653E-03	1.478E-03
15.8	1.658E-03	1.483E-03
15.9	1.663E-03	1.487E-03
16.0	1.667E-03	1.491E-03
16.1	1.671E-03	1.494E-03
16.2	1.675E-03	1.497E-03
16.3	1.678E-03	1.500E-03
16.4	1.680E-03	1.502E-03
16.5	1.683E-03	1.504E-03
16.6	1.685E-03	1.506E-03
16.7	1.687E-03	1.508E-03
16.8	1.689E-03	1.510E-03
16.9	1.690E-03	1.511E-03
17.0	1.691E-03	1.512E-03
17.1	1.693E-03	1.513E-03
17.2	1.694E-03	1.514E-03
17.3	1.695E-03	1.515E-03
17.4	1.695E-03	1.516E-03
17.5	1.696E-03	1.516E-03
17.6	1.697E-03	1.517E-03
17.7	1.697E-03	1.518E-03
17.8	1.698E-03	1.518E-03
17.9	1.698E-03	1.518E-03
18.0	1.699E-03	1.519E-03
18.1	1.699E-03	1.519E-03
18.2	1.700E-03	1.519E-03
18.3	1.700E-03	1.520E-03
18.4	1.700E-03	1.520E-03
18.5	1.700E-03	1.520E-03
18.6	1.701E-03	1.520E-03
18.7	1.701E-03	1.520E-03
18.8	1.701E-03	1.521E-03
18.9	1.701E-03	1.521E-03
19.0	1.701E-03	1.521E-03
19.1	1.701E-03	1.521E-03
19.2	1.701E-03	1.521E-03
19.3	1.702E-03	1.521E-03
19.4	1.702E-03	1.521E-03
19.5	1.702E-03	1.521E-03
19.6	1.702E-03	1.521E-03
19.7	1.702E-03	1.521E-03
19.8	1.702E-03	1.521E-03
19.9	1.702E-03	1.522E-03
20.0	1.702E-03	1.522E-03



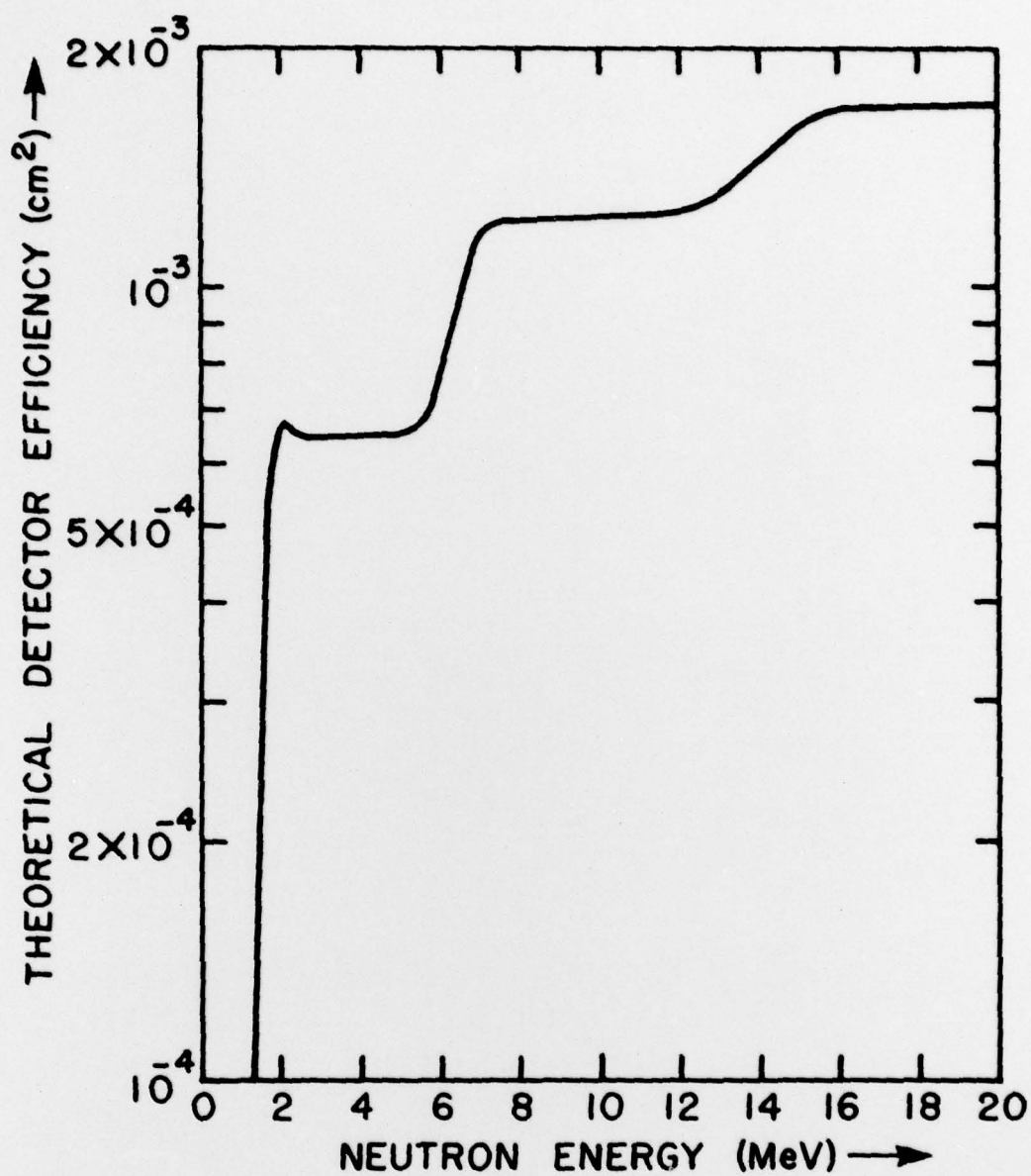


Figure 1 : Theoretically determined efficiency of the depleted uranium-238 fission chamber.



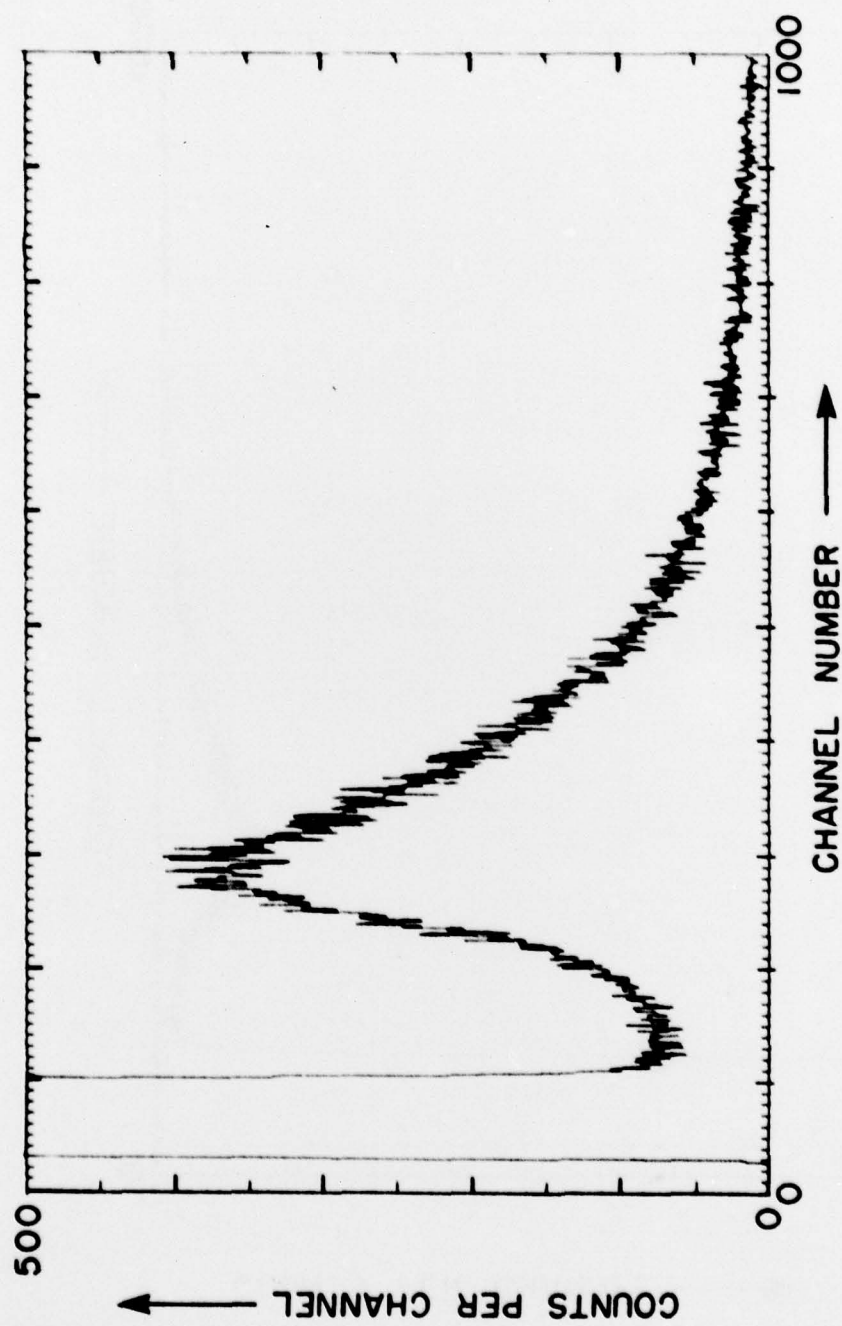


Figure 2 : Observed gross pulse-height spectrum induced by the californium-252 source.

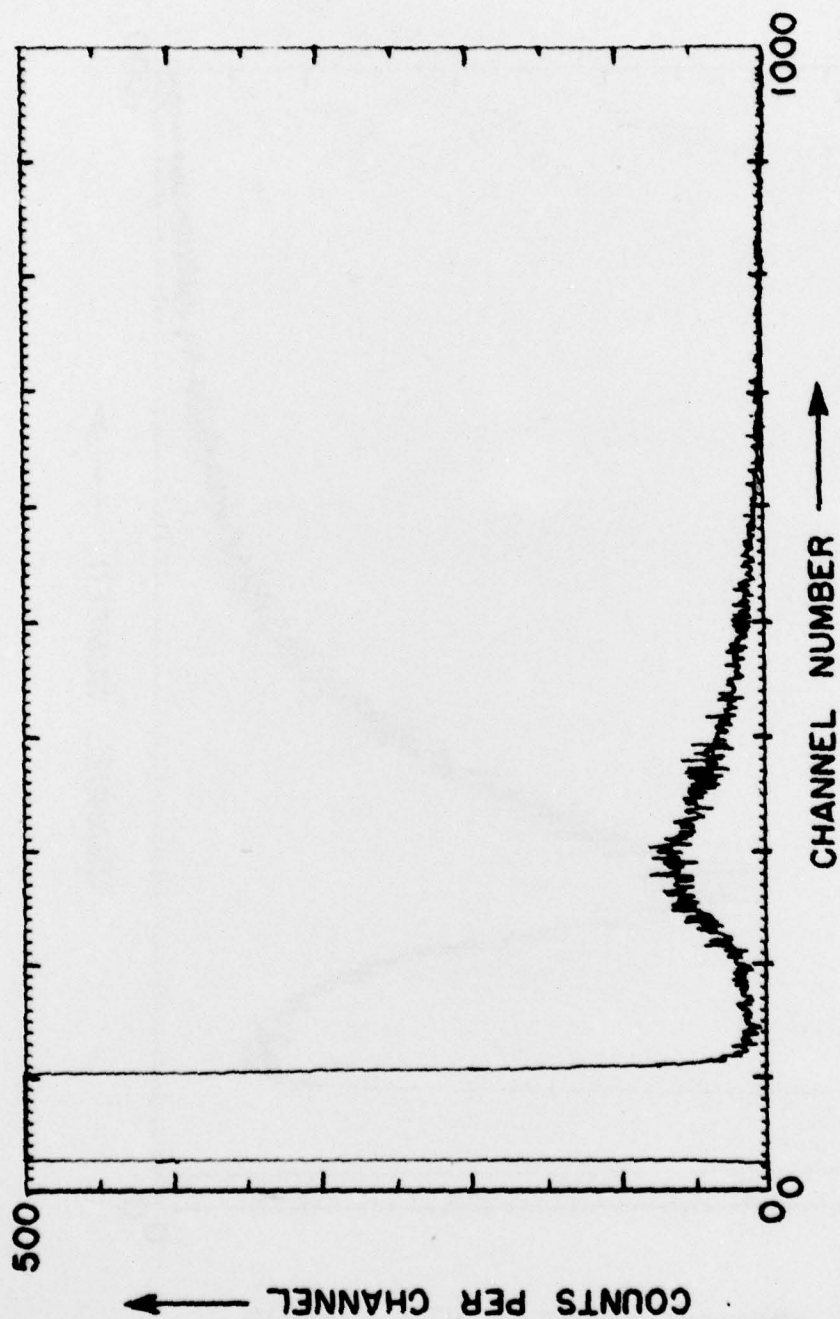


Figure 3 : Background pulse-height spectrum obtained with a paraffin cylinder interposed between the californium-252 source and the fission chamber.

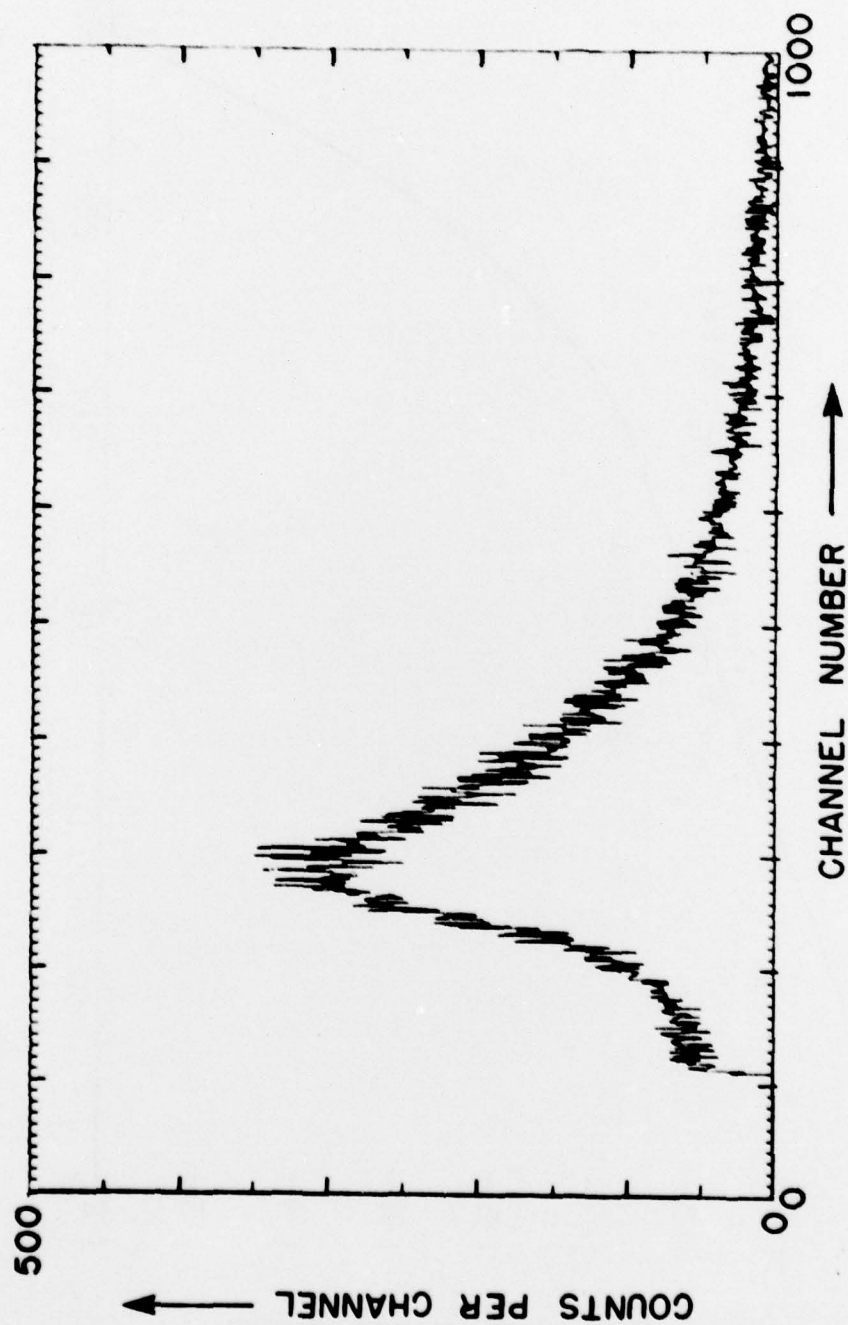


Figure 4 : Net foreground pulse-height spectrum induced by the californium-252 source.

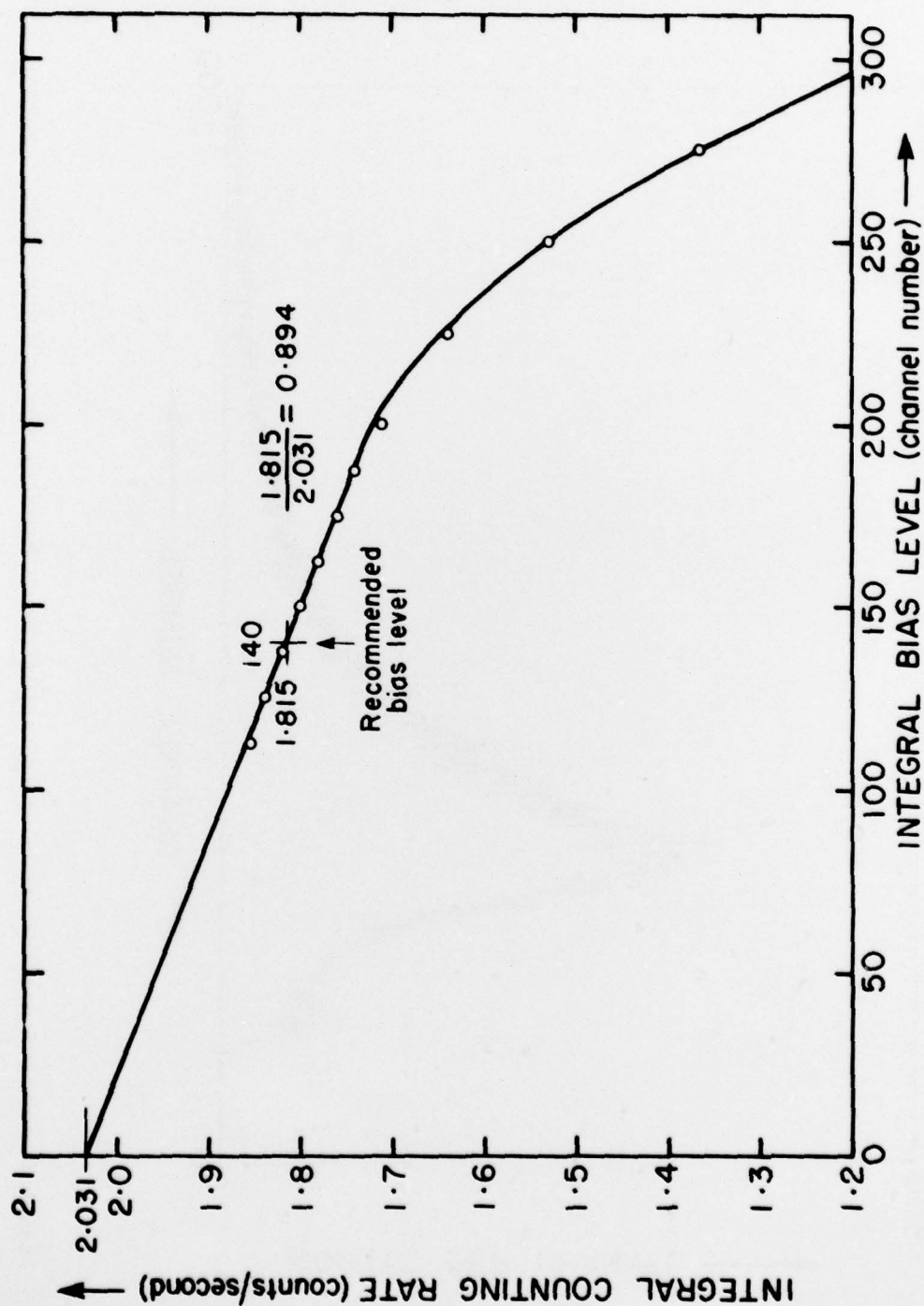


Figure 5 : Integral pulse-height spectrum induced by the calcium-252 source.



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